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# Impact-parameter-dependent multifragmentation of C-60 in charge-changing collisions with 2-MeV C<sup>+</sup> ions

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# Impact-parameter-dependent multifragmentation of $C_{60}$ in charge-changing collisions with 2-MeV $C^+$ ions

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Multifragmentation of  $C_{60}$  was investigated for 2 MeV  $C^+$  projectile ions by means of a time-of-flight triple coincidence technique. Fragment ions and secondary electrons were measured simultaneously in coincident with charge-selected outgoing projectile particles scattered into forward angles smaller than 1.0 mrad. Fragment ion distributions were found to change remarkably with respect to the scattering angle  $\theta$ . Relationships between  $\theta$  and the impact parameter  $b$  measured from the center of  $C_{60}$  were calculated classically. We found evidently that small-size fragment ions are produced only from cage-penetration collisions, while intact parent ions and their daughter ions are created in collisions nearby but outside a  $C_{60}$ . Furthermore, charge state distributions of prefragmented  $C_{60}^{r+}$  ions and correlated fragment ion pairs were both found to be essentially independent of  $\theta$  when plotted for individual  $C_m^+$  ions of fixed size. These results indicate that the production of a size-fixed specific ion may be restricted to a certain impact parameter region in which equivalent electronic energies are deposited irrespectively of  $\theta$ . This was confirmed also from our calculation of the energy deposition as a function of  $b$  carried out using the local density approximation.

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## I. INTRODUCTION

In the past decade, fullerene molecules have attracted considerable attention as a collision partner providing information about properties of matter lying between atoms and solids [1]. Among many experimental results on ionization and fragmentation of  $C_{60}$  obtained to date by various primary probes of charged particles and photons [1–18], it is noteworthy that the  $C_{60}$  molecule has remarkable high stability against Coulomb repulsive force. For instance, evidence for the existence of highly ionized intact ions like a  $C_{60}^{12+}$  is reported in recent experimental work using an intensive femtosecond laser [4]. On the other hand, charged particle bombardment on  $C_{60}$  tends to result in molecular fragmentation arising from instability due to Coulomb repulsive force and to internal excitation energy. The former mechanism is important in collisions with slow highly charged ions (SHCI), since multiply charged prefragmented ions  $C_{60}^{r+}$  are easily produced via multiple electron capture taking place at rather large impact parameters [6]. In such collisions, the internal excitation energy is supposed to be small. The latter mechanism, i.e., instability due to high internal excitation, becomes significant in fast ion collisions, where a large amount of inelastic energy  $E_d$  is deposited into a  $C_{60}$  [7–18]. As the total amount of  $E_d$  is supposed to depend on the impact parameter  $b$  between collision partners, it is important to know the relationship between  $E_d$  and  $b$ . Nevertheless, no such investigations have been carried out so far except for SHCI experiments [19–23] of scattering angle dependent electron capture collisions. Among these investigations, Cederquist *et al.* discussed in detail the relationship between fragmentation and  $b$  [23].

In the present work, fast ion induced  $C_{60}$ -multifragmentation is investigated at scattering angles from 0 to 1.0 mrad. At each scattering angle we carried out simultaneous measurements of fragment ions, the number of secondary electrons, and charge-selected outgoing projectile particles. This triple coincidence technique allows us to obtain information about (i) correlation between fragment ion pairs and (ii)  $r$  distributions of  $C_{60}^{r+}$  in correlation with a  $C_m^+$  ion of any cluster size  $m$ . As the scattering angle  $\theta$  is closely related to the impact parameter  $b$  in collisions, all the  $\theta$ -dependent data can be interpreted in terms of  $b$  as described in the following sections.

## II. EXPERIMENT

The experiment was performed at the QSEC heavy ion facility of Kyoto University. The experimental method and apparatus are described elsewhere [14,16,17], so that only a brief outline is given below.

A beam of 2.0 MeV  $C^+$  ions provided from a 1.7 MV tandem accelerator was carefully collimated to smaller than 0.1 mm in diameter by two-dimensional knife-edged slits and was charge purified with a magnetic charge-selector before entering a collision chamber. A gas phase  $C_{60}$  target was produced by sublimation of high-purity (99.98%) powder at 550 °C. A base pressure of the target chamber was kept below  $5 \times 10^{-6}$  Pa. After collisions with the  $C_{60}$  target, outgoing projectiles were charge separated horizontally by an electrostatic deflector and detected by a movable semiconductor detector (SSD) located 1 m downstream of the collision chamber. The scattering angle  $\theta$  was resolved by placing a hole slit of 0.5 mm in diameter in front of the SSD. An acceptance angle of the detector was 0.5 mrad.

Positive fragment ions and secondary electrons were extracted into opposite directions by an electric field of

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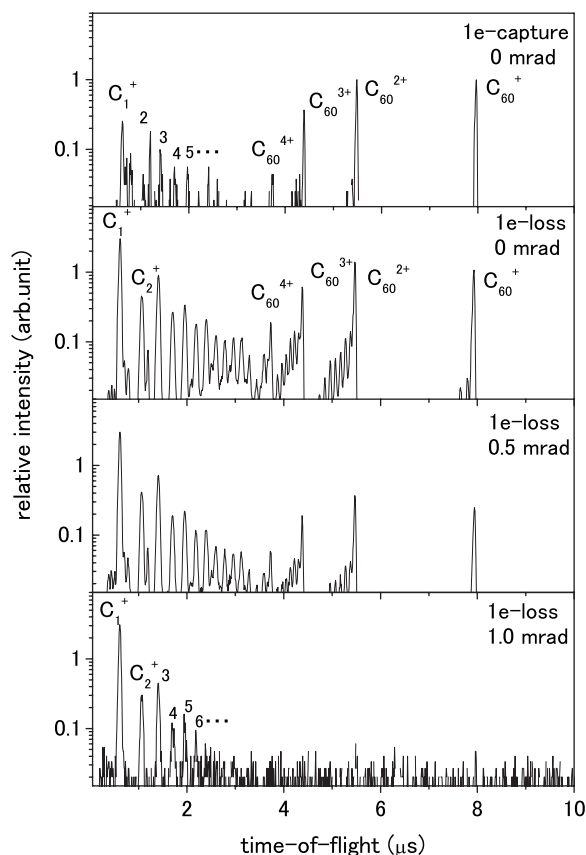


FIG. 1. Time-of-flight spectra of fragment ions produced in 1e-capture and 1e-loss collisions of 2 MeV  $C^+$  measured at scattering angles of  $\theta=0, 0.5$ , and  $1.0$  mrad.

615 V/cm applied perpendicular to the incident beam axis. Fragment ions were detected by a two-stage multichannel plate with a front voltage of  $-4.6$  kV. The mass distribution of product ions was measured by a time-of-flight method. Secondary electrons were detected by a PIPS type detector biased at  $+30$  kV, enabling us to obtain the number of secondary electrons  $n_e$  emitted simultaneously. Detection of fragment ions and secondary electrons was carried out in coincidence with charge-and-scattering angle resolved scattered ions. We applied this triple coincidence technique to single electron loss and capture collisions.

### III. RESULTS AND DISCUSSION

Figure 1 shows time-of-flight spectra of fragment ions measured for single electron capture (1e-capture) and single electron loss (1e-loss) collisions of 2.0 MeV  $C^+$  with  $C_{60}$ . Data were taken at three scattering angles of  $\theta=0\pm0.25, 0.5\pm0.25$ , and  $1.0\pm0.25$  mrad. It was found that 1e-capture events were considerably smaller than 1e-loss and the difference was more than two orders of magnitude. Also, in 1e-capture collisions almost no peak intensities were found at  $\theta=0.5$  and  $1.0$  mrad. One can see that spectral profiles in 1e-loss collisions change remarkably depending on the scattering angle. Ionized parent ions  $C_{60}^{r+}$  and their fullerene-like daughter ions decrease rapidly as  $\theta$  increases and they nearly vanish at  $1.0$  mrad.

In order to know the relationship between the scattering angle  $\theta$  and the impact parameter  $b$  measured from the center of  $C_{60}$ , a simulation calculation has been done as follows. The calculation was performed for 100 000 incident particles with randomly generated projectile positions  $(x,y)$  with  $x^2+y^2=b^2$ , where the plane  $(x,y)$  is perpendicular to the incident beam direction. Molecular orientation of  $C_{60}$  was also generated randomly with respect to the incident beam direction. For each value of  $b$  impact parameters  $p$ 's between the incident ion and 60 carbon atoms were calculated individually. Here we calculated only binary collisions between the incident ion and the closest carbon atom because multiple collision events inside  $C_{60}$  are negligibly small. Furthermore, we restricted our calculation to a range of  $p\leq 4$ , corresponding to about twice the atomic carbon radius [24], since electron loss and capture collisions are unlikely to occur at  $p>4$  in fast collisions [25]. The scattering angle  $\Theta$  in the center-of mass (c.m.) frame was calculated classically as

$$\Theta = \pi - 2p \int_{r_c}^{\infty} r^{-2} \left( 1 - \frac{p^2}{r^2} - \frac{V(r)}{E_{c.m.}} \right)^{-1/2} dr, \quad (1)$$

where  $r$  is the internuclear distance,  $p$  the impact parameter,  $E_{c.m.}$  the incident energy in the c.m. frame,  $r_c$  the closest approach distance, and  $V(r)$  the interaction potential for which we used the ZBL-type universal potential [26]. As the collision partners are identical and an inelastic energy loss is estimated to be negligibly small compared to the incident energy [27], the scattering angle  $\theta$  in the laboratory frame is approximated by  $\theta=\Theta/2$ .

Calculated yields of scattering into  $\theta=0, 0.5$ , and  $1.0$  mrad are plotted in Fig. 2 as a function of  $b$ . The number density of target carbon atoms is also depicted in the upper figure. It is found that scattering into non-zero angles ( $\theta \neq 0$ ) is essentially limited to  $b \leq 7$ , i.e., cage penetration collisions. Together with the results from Fig. 1, we find that small fragment ions like  $C_{1,2,3}^+$  are produced in these cage penetration collisions, while parent ions  $C_{60}^{r+}$  are produced outside or in a peripheral area of  $C_{60}$ .

Present results coincide reasonably with those obtained in SHCI experiments [5,6]. In our fast ion collisions, fission into two charged fragment ions such as  $C_{60}^{4+} \rightarrow C_{58}^{3+} + C_2^+$  was found to occur scarcely and multifragmentation is dominant. Examples of emission distributions of fragment ions correlated with  $C_1^+$  and  $C_3^+$  are shown in Fig. 3. One can see that the fragment yields decrease with increasing number of carbon atoms, and, as expected, the yield correlated with  $C_1^+$  decreases more rapidly in comparison with  $C_3^+$ . An important result is that the fragment distribution is less sensitive to the scattering angle.

Figure 4 shows charge state distributions of prefragmented  $C_{60}^{r+}$  ions obtained for fixed-sized fragment ions of  $C_n^+$  ( $n=1,3,5$ ). The  $r$ -distribution shows again almost the same profiles independently of the scattering angles. For comparison, data obtained for  $C_1^+$  in 1e-capture collisions at  $\theta=0$  are also plotted in the uppermost figure. One can see that the distribution profile is also the same as 1e-loss collisions. Present results of  $\theta$ -independency both for mass distributions (Fig. 3) and  $r$ -distributions indicate clearly that a

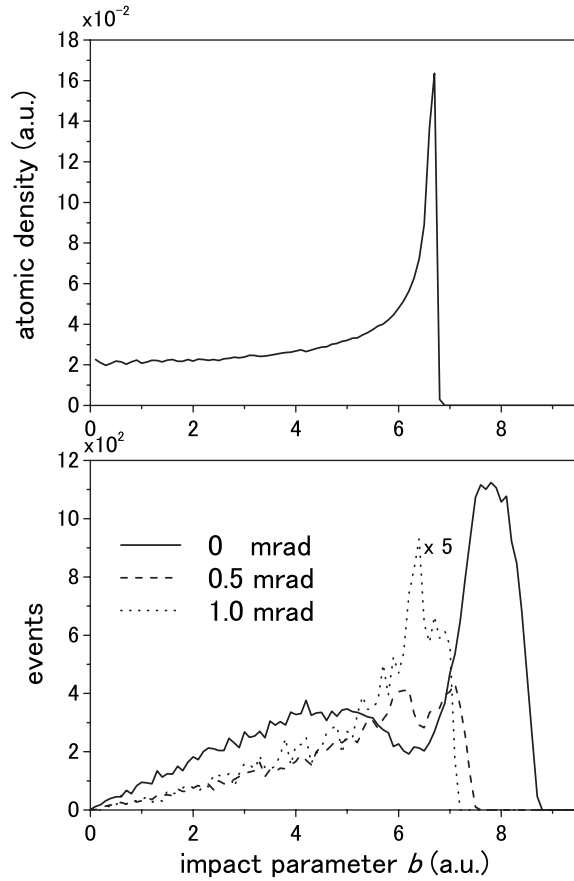


FIG. 2. Calculated scattering yields for  $\theta=0, 0.5$ , and  $1.0$  mrad plotted as a function of the impact parameter  $b$  measured from the center of  $C_{60}$ .

specific fragment ion is produced in equivalent impact parameter collisions independently of the scattering angle. The similar conclusion was also derived in our previous work using  $2 \text{ MeV Si}^{2+}$  ions [17].

In order to know the most probable impact parameters at which fixed-size specific fragment ions are produced preferentially, we calculated the electronic energy deposition  $E_d$  as a function of the impact parameter  $b$ , as described below. The calculation was done by using the local density approximation (LDA) [28–30]. The mean electronic energy loss of an incident particle (charge  $q$  and velocity  $v$ ) in an electron gas is given by

$$-\frac{dE}{dx}(r) = \frac{4\pi q^2}{v^2} \rho(r) L(\rho(r), v), \quad (2)$$

where  $r$  the position of the incident ion measured from the  $C_{60}$  center,  $\rho(r)$  the electron density, and  $L$  the stopping number. The electron density is given by the following analytical expression [11,31].

$$\rho(r) = 0.146 \exp\left(\frac{-(r-6.6)^2}{2.7}\right). \quad (3)$$

As the velocity of  $2 \text{ MeV C}_1^+$  ions is larger than the local Fermi velocity defined as  $v_F(r) = [3\pi^2 \rho(r)]^{1/3}$ ,  $L$  is obtained by

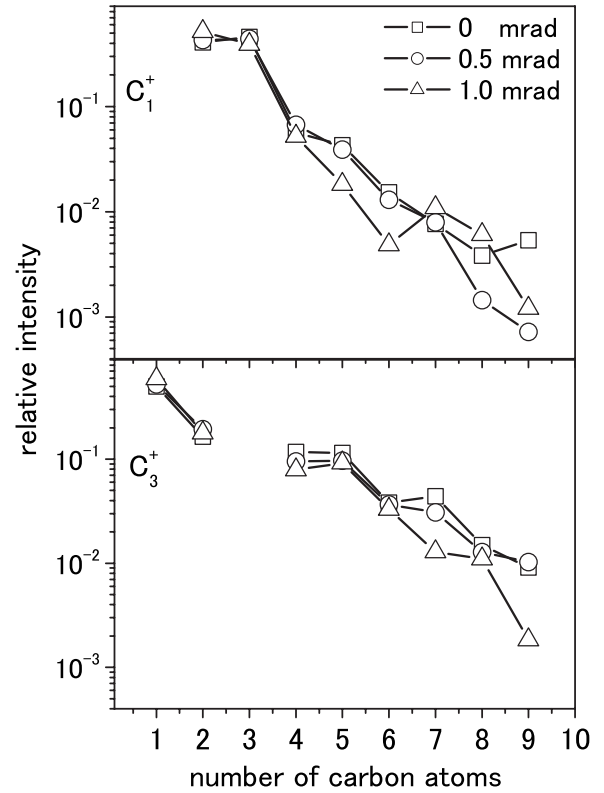


FIG. 3. Relative intensity of fragment ions  $C_m^+$  produced simultaneously with  $C_1^+$  (upper) and  $C_3^+$  (lower) as a function of the cluster size  $m$  obtained for  $1e$ -loss collisions of  $2 \text{ MeV C}^+$  at scattering angles of  $\theta=0, 0.5$ , and  $1.0$  mrad.

$$L(\rho(r), v) = \ln\left(\frac{2v^2}{\omega_p(r)}\right) - \frac{3}{5}\left(\frac{v_F(r)}{v}\right)^2, \quad (4)$$

with the plasma frequency  $\omega(r) = [4\pi\rho(r)]^{1/2}$ . The mean energy deposition  $E_d$  for a certain impact parameter  $b$  is obtained by a linear integral of Eq. (2) along the beam trajectory as

$$E_d(b) = \frac{4\pi q^2}{v^2} \int_{-\infty}^{\infty} \rho(r) L(\rho(r), v) dz. \quad (5)$$

In calculations, we used instead of  $q$  an effective charge obtained from [32].

Calculated results of  $E_d$  are presented in Fig. 5 as a function of  $b$ . The value of  $E_d$  reaches about  $800 \text{ eV}$  in the peripheral area of  $C_{60}$ , and its average value is about  $600 \text{ eV}$ . As we discussed in detail in [7,16,17], the total energy deposition  $E_d$  is shared by ionization and internal excitation with a certain partition rate. Therefore once the degree of ionization ( $r$ ) is known, the value of  $E_d$  may be deduced separately for each fragment ion. As for partition rates, we used theoretical values reported for other collision systems of  $0.2\text{--}10^4 \text{ keV H} + \text{H}_2\text{O}$  [33] and  $1.4 \text{ MeV/amu U}^{32+} + \text{Ne}$  [34]. According to these papers, about 20% of  $E_d$  is spent for internal excitation and 80% for ionization energy [33], and 75% of the ionization energy is carried away by ionized electrons as their kinetic energies [34]. The rest of the ionization energy ( $E_d \times 0.8 \times 0.25 = E_d \times 0.2$ ) equals a sum of ionization

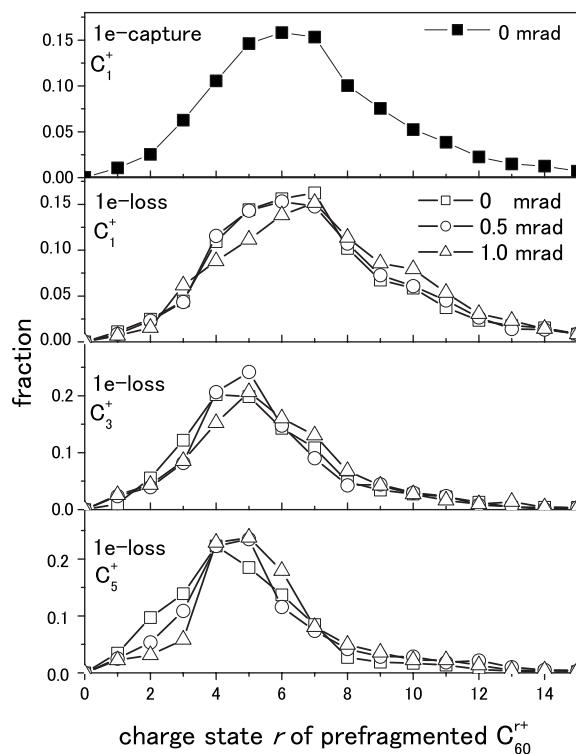


FIG. 4. Charge state distribution correlated with specific fragment ions  $C_m^+$  in single electron loss collisions of 2 MeV  $C^+$  at scattering angles of  $\theta=0, 0.5$ , and  $1.0$  mrad.

potentials of all the ionized electrons. Hence  $E_d$  is related to the degree of ionization as

$$E_d \times 0.2 = \sum_{i=1}^r I_i, \quad (6)$$

where  $I_i$  is the  $i$ th ionization potential given by  $I_i=3.77+3.82i$  [3]. As for  $C_1^+$ , for instance,  $E_d$  is calculated to be 665 eV by using an average value of  $\bar{r}=7$  obtained from  $r$ -distributions as shown in Fig. 4. In this way, we obtained total energy depositions  $E_d$  for each fragment ions and plotted them in Fig. 5. We find that  $C_1^+$  is predominantly produced at impact parameters of 5–7 a.u. (cage penetration) and medium size ions  $C_m^+$  ( $m=5\sim 8$ ) are produced at about 8 a.u. These results are consistent with those obtained from scattering angle measurements described above.

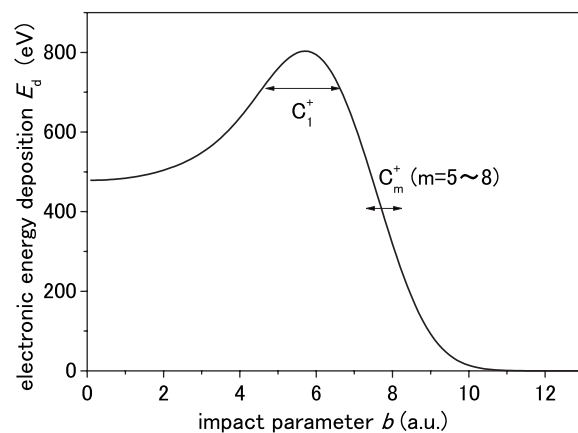


FIG. 5. Electronic energy deposition  $E_d$  into a  $C_{60}$  by 2 MeV  $C^+$  as a function of the impact parameter  $b$ . The most probable impact parameters for the production of  $C_1^+$  and  $C_m^+$  are also shown by arrows.

In summary, collision-induced  $C_{60}$  fragmentation following charge exchange of 2 MeV  $C^+$  ions scattered into small forward angles has been investigated. It was found that the charge-state distribution of prefragmented  $C_{60}^{r+}$  ions correlated with fixed-size fragment ions  $C_m^+$  is essentially independent of both scattering angle and projectile final charge. This characteristic was also found for mass distributions of correlated fragment ion pairs. It implies evidently that fixed-size fragment ions are produced in equivalent impact parameter collisions independently of the projectile scattering angle.

We attempted calculations to determine the most probable impact parameters for the production of specific fragment ions by using an energy partition model. Present experimental results can be reasonably interpreted by our simple calculations. It should be noted, however, that the charge-state distributions are spread widely and it needs more accurate statistical analysis to achieve more realistic understanding of fragmentation mechanism.

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